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Grain shape dependence of the thermal expansion coefficient of a binary composite

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Abstract : Multiple scattering theory has been used to calculate the effective thermal expansion coefficient of a binary composite whose phases are made up of spheroidal grains. Explicit expressions for disc, sphere and needle shaped grains are presented. In absence of any experimental data, the formulae obtained are discussed in case of Al/Al_2O_3 composite.

Keywords : Binary composite, thermal expansion coefficient, grain shape dependence

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1. Introduction

This is a sequel to a previous communication [1] in which Ballabh *et al* used the multiple scattering theory (mst) to calculate the effective thermal expansion coefficient α^* of a binary composite composed of spherical grains. In the present work, we have applied the mst to predict how α^* depends on the grain shape of the constituent phases of a binary composite.

Previous works on the determination of α^* were confined to calculate the strain field within a spherical inclusion and then to evaluate its average either from the constitutive relation (eq. 1) or from a similar relation in terms of complementary energy [2]. Expression for α^* thus obtained for a binary composite depends on the thermal expansion coefficient of both phases and the effective bulk modulus (K^*) of the composite. These works are briefly reviewed in Refs. [3] and [4].

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Levin [5] and Rosen and Hashin [6] derived expressions for effective elastic constants and α^* using the field solution within a spherical inclusion in a homogeneous matrix. α^* does not depend explicitly on the surrounding matrix but depends on K^* which, however, depends on the surrounding matrix.

The usual procedure [4] to calculate the grain shape dependence of α^* is to develop a theory for α^* which, as stated, earlier, depends on K^* and then to use another formalism which gives the grain shape dependence of K^*. It is usually overlooked whether the two theories are based on the same assumptions or not.

In the next section, formal expressions for α^* and the effective elastic constants (C^*) have been derived in terms of a single grain scattering matrix using multiple scattering theory. Using a self-consistent scheme, explicit expressions for α^* and C^* of a binary composite containing spheroidal grains are evaluated in Section 3. Finally in Section 4, the formulae obtained are discussed.

2. Formal expressions for α^* and C^*

In case of a homogeneous material, the thermal expansion coefficient (TEC) α_{kl} is defined by

$$\sigma_{ij} = C_{ijkl}\,\varepsilon_{kl} - C_{ijkl}\,\alpha_{kl}\,\phi, \tag{1}$$

where σ, ε and C are respectively, the stress tensor, the strain tensor and elastic constant tensor. ϕ is the increase in temperature from some reference temperature.

For an inhomogeneous material, σ, ε, C and α are position dependent *i e.* $\sigma = \sigma(\boldsymbol{r})$; $\varepsilon = \varepsilon(\boldsymbol{r})$ *etc*. But we shall assume that the material is in thermal equilibrium *i.e.* ϕ is independent of $\boldsymbol{r}$.

Similar to eq. (1), the effective thermal expansion coefficient α^* is defined in terms of the effective elastic constants C^* by the relation

$$<\sigma> = C^* <\varepsilon> - C^*\alpha^*\phi, \tag{2}$$

where < > denotes ensemble averaging which in the case of a composite with nonspherical grains includes both volume averaging and shape orientation averaging.

To calculate C^* and α^* we have to determine the strain field solution within the inhomogeneous material. For this, we consider the inhomogeneous material of elastic constant $C(\boldsymbol{r})$ and TEC $\alpha(\boldsymbol{r})$ as a homogeneous and isotropic medium of elastic constant C^0 and TEC α^0 with the inhomogeneity $\delta C(\boldsymbol{r})$ and $\delta\alpha(\boldsymbol{r})$ superposed on it such that

$$C(\boldsymbol{r}) = C^0 + \delta C(\boldsymbol{r}) \tag{3}$$

and

$$\alpha(\boldsymbol{r}) = \alpha^0 + \delta\alpha(\boldsymbol{r}). \tag{4}$$

Now, using the mechanical equilibrium condition

$$\nabla\cdot\sigma = 0 \tag{5}$$

the strain field solution is obtained [1] by superposing the relevant fields scattered by the inhomogeneities within the medium characterised by C^0 and α^0. The equilibrium strain field solution in operator form is obtained as

$$\varepsilon = \varepsilon^0 - \alpha^0\phi + G\delta C(\varepsilon - \alpha\phi) - GC^0\delta\alpha\phi. \tag{6}$$

In eq. (6), $(\varepsilon^0 - \alpha^0\phi)$ is the strain due to homogeneous and isotropic scattering medium at equilibrium. The remaining parts on the right hand side, represent the strain scattered by the inhomogeneity δC and $\delta\alpha$. G is the strain Green function for the homogeneous medium and for spheroidal grain is given by [7–9]

$$G = -AS^0, \tag{7}$$

where $S^0 = (C^0)^{-1}$ and A is a fourth rank tensor which has an isotropic and a direction dependent part. The details of A is given in Appendix I.

Now, iteration of eq. (6) gives

$$\varepsilon = (I + GT)(\varepsilon^0 - \alpha^0\phi - GC^0\delta\alpha\phi) - GT\alpha\phi, \tag{8}$$

where $$I = I_{ijkl} = \frac{1}{2}\left(\delta_{ik}\delta_{jl} + \delta_{il}\delta_{jk}\right) \tag{9}$$

and $$T = \delta C + \delta CGT = \delta C(I - G\delta C)^{-1}. \tag{10}$$

T is formally called the 'transition matrix' because it completely specifies 'transition' out of the field ε^0. The term matrix arises from the use of 'T' in quantum mechanics but here 'T' represents a fourth rank tensor.

In the absence of ϕ, ε must satisfy the boundary condition $\varepsilon^0 = <\varepsilon>$. Thus

$$\langle I + GT\rangle = I \tag{11}$$

$$\text{or,}\ \ \langle GT\rangle = 0 \tag{12}$$

$$\text{or,}\ \ \langle q\rangle = I, \tag{13}$$

where $$I + GT = q. \tag{14}$$

Using eqs. (1), (2) and (11) – (14), we get the following formal expressions for C^* and α^*

$$C^* = \langle Cq\rangle = C^0 + \langle T\rangle \tag{15}$$

and $$C^*\alpha^* = (C^0 - C^*)\langle qGC^0\delta\alpha + GT\alpha\rangle + \langle C\alpha q\rangle. \tag{16}$$

For a general homogeneous media, the T-matrix is impossible to calculate except for a very few simple cases. In the single grain scattering approximation, T is written as

$$T = \sum_i t^i = \tau, \tag{17}$$

where the summation is over i (*i.e.* all grains) and t^i is the single grain scattering matrix which in analogy with total T matrix, can be written as

$$t^i = \delta C^i + \delta C^i G t^i. \tag{18}$$

With a single grain scattering approximation, eqs. (15) and (16) become

$$C^* = \langle CQ \rangle = C^0 + \langle \tau \rangle \tag{19}$$

and
$$C^*\alpha^* = (C^0 - C^*)\langle QGC^0\delta C + G\delta CQ\alpha \rangle + \langle C\alpha Q \rangle, \tag{20}$$

where
$$Q = I + G\tau \tag{21}$$

with $< Q > = I$ [from eq. (11)]. Eqs. (19) and (20) are formal expressions for C^* and $C^*\alpha^*$ under the single grain scattering approximation.

3. Self-consistent approximation

To evaluate explicit expressions for C^* and α^* from eqs. (19) and (20) we have to specify C^0. In the self-consistent approximation C^0 is chosen in such a way that $C^0 = C^*$. With this choice, we get from eqs. (19) – (21).

$$C^* = \langle CQ \rangle \tag{22}$$

$$\text{or,} \ \langle \delta CQ \rangle = 0 \tag{23}$$

and
$$C^*\alpha^* = \langle C\alpha Q \rangle \tag{24}$$

with
$$\langle Q \rangle = I. \tag{25}$$

Eq. (23) together with eq. (25) gives the following expression for the effective bulk (K^*) and shear modulus (μ^*) of a binary composite (details are given in Appendix II)

$$K^* = K_2 - \upsilon_1(K_2 - K_1)\overline{(Q_{11} + 2Q_{12})}_1 \tag{26}$$

and
$$\mu^* = \mu_2 - \upsilon_1(\mu_2 - \mu_1)\overline{(2Q_{44})}_1, \tag{27}$$

where the bar denotes shape average. The subscripts 1 and 2 refer to phases 1 and 2 respectively. υ_1 is the volume fraction of phase 1. In deriving eqs. (26) and (27), phase 1 is taken as inclusion and phase 2 as the matrix. Similar expression will be obtained if the inclusion and the matrix are interchanged. But if the inclusion and matrix phases are interchanged, the formulae obtained will be different from the eqs. (26) and (27). This is due to the assymmetric nature of the theory introduced by conditions [eqs. (23) and (25)]. Thus, choice of matrix and inclusion phase is important. It is important to note that K^* and μ^* depend only on the grain shape of the inclusion.

If we now substitute the expressions of $\overline{(Q_{11} + 2Q_{12})}_1$ and $\overline{(2Q_{44})}_1$ from Appendix II in eqs. (26) and (27), we get two simultaneous equations involving K^* and μ^*. These equations can be numerically solved to obtain K^* and μ^*. In the special cases of a sphere, needle and disc eqs. (26) and (27) reduce to the general expression

$$\frac{C^* - C_1}{C_2 - C_1} = \frac{\upsilon_2}{\left[1 + \frac{\upsilon_1(C_2 - C_1)}{C_1 + F}\right]}, \tag{28}$$

where C denotes either K or μ and F is a function of K_1, K_2, μ_1, μ_2, K^* and μ^*. The specific forms of F for sphere, needle and disc are given below.

Sphere :

For K^*, $\quad F = \frac{4}{3}\mu^*.$

For μ^*, $\quad F = \dfrac{\mu^*(9K^* + 8\mu^*)}{6(K^* + 2\mu^*)}.$

Needle :

For K^*, $\quad F = \frac{1}{3}\mu_1 + \mu^*.$

For μ^*, $\quad F = \dfrac{5}{M} - \mu_1,$

where $\quad M = \dfrac{1}{(3K_1 + \mu_1 + 3\mu^*)} + \dfrac{2(2\mu_1 + \mu^* + g)}{(\mu_1 + \mu^*)(\mu_1 + g)}$

with $\quad g = \dfrac{\mu^*\left(K^* + \frac{1}{3}\mu^*\right)}{\left(K^* + \frac{7}{3}\mu^*\right)}.$

Disc :

For K^*, $\quad F = \frac{4}{3}\mu_1.$

For μ^*, $\quad F = \dfrac{\mu_1(9K_1 + 8\mu_1)}{6(K_1 + 2\mu_1)}.$

Eq. (28) is identical to the expression given in the review article of Watt *et al* [10] who concluded, after simplifying several self-consistent results for two phase composite, that all the self-consistent results can be put in the form of eq. (28).

To get the expression for α^*, we use eqs. (23) and (24) and obtain for a two phase composite

$$\alpha^* = \alpha_1 + \frac{(\alpha_1 - \alpha_2)\left(\frac{1}{K^*} - \frac{1}{K_1}\right)}{\left(\frac{1}{K_1} - \frac{1}{K_2}\right)} \tag{29}$$

which is a well known result.

Thus the grain shape dependence of α^* comes through K^* which as discussed earlier can be calculated from eqs. (26) and (27) numerically.

4. Discussion

The formulae developed in Section 3, suggest that it is possible to calculate the grain shape dependence of α^* if that of K^* is known. In deriving the grain shape dependence of K^* (eq. 26), phase 1 is taken as an inclusion and phase 2 as the matrix. Excepting the case of spherical grains, the formula is not symmetrical if the inclusion and the matrix are interchanged. Similar results are also applicable for μ^*. Thus K^* and hence α^* depend on the choice of the matrix or inclusion phase. As discussed in Section 3, this is due to the asymmetric nature of the present formalism.

Finally, the formulae obtained in Section 3 are applied in case of an Al_2O_3/Al composite [11]. The input data are given in Table 1. The results obtained for sphere, needle

Table 1. Input data for Al_2O_3/Al composite at 100°C. Data taken from Elomari *et al* [11].

Material	Shear modulus in GPa	Bulk modulus in GPa	CTE in 10^{-6} per °C
Al	26.5	78.9	23.5
Al_2O_3	162.25	270.3	6.4

and disc shaped grains of the inclusion are shown graphically in Figure 1. The graph shows that for spherical and needle shaped grains the results are almost identical. For disc the

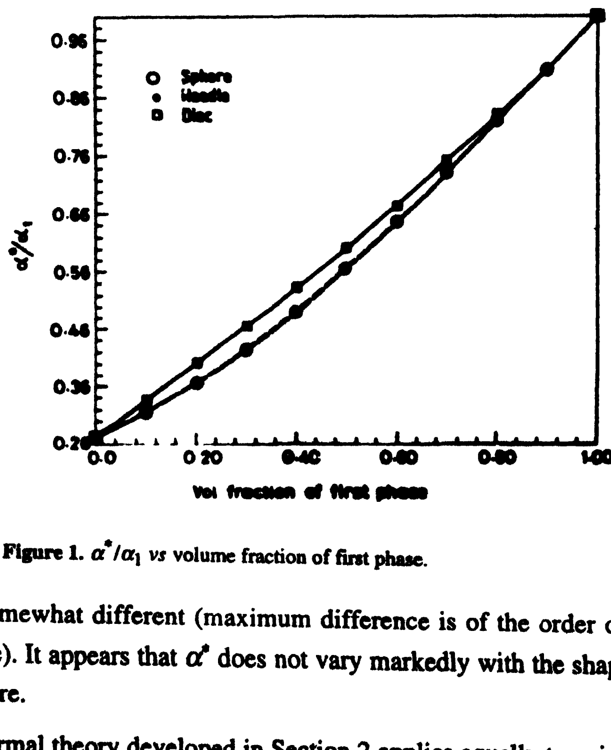

Figure 1. α^*/α_1 *vs* volume fraction of first phase.

results are somewhat different (maximum difference is of the order of 10% from that of spherical case). It appears that α^* does not vary markedly with the shapes of the inclusions considered here.

The formal theory developed in Section 2 applies equally to anisotropic composites, but the averaging procedure will be different. In addition to volume averaging and phase shape orientation averaging, direction averaging of grains will have to be performed. Anisotropic composites specifically fiber reinforced composites find wide application in material science. Work in this direction is in progress. Apart from these, the formal theory can also be extended to polycrystals where the calculation will be a bit involved.

The present formalism cannot take into account the size effect of inclusion or matrix grains. Further, the present theory treats a composite as a model particulate system where matrix and inclusion grains are distributed randomly as separate phases and the whole composite is isotropic in nature. But in real composites, the picture may be different where the composite is made up of regions where a number of inclusion grains is surrounded by a single matrix grain. In that case, the sample will be anisotropic and the strain distribution will be quite different. The theory presented here needs modification in that case.

In the absence of any experimental data, it is not possible to compare the present results with experiment. However, it will be immensely useful if the present work can stimulate experiments designed to study the grain shape dependence of different physical properties of composites.

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Appendix I

The fourth rank tensor A in eq. (7) has the following symmetry. (In Ref. [8], A is denoted by S and in Ref. [9] by W^{-1}).

$$A_{11} = A_{22}; A_{33}; A_{12} = A_{21}; A_{13} = A_{23}; A_{31} = A_{32};$$

$$A_{44} = A_{55}; A_{66}; A_{11} - A_{12} - 2A_{66} = 0.$$

Explicitly the components of A are given by

$$A_{11} = P\left(\pi - \frac{La^2}{4}\right) + RI, \tag{30}$$

$$A_{33} = P\left(\frac{4\pi}{3} - \frac{2c^2L}{3}\right) + R(4\pi - 2I). \tag{31}$$

$$A_{12} = P\left(\frac{\pi}{3} - \frac{a^2L}{12}\right) - RI, \tag{32}$$

$$A_{13} = P\frac{c^2L}{3} - RI, \tag{33}$$

$$A_{31} = P\frac{a^2L}{3} - R(4\pi - 2I), \tag{34}$$

$$A_{44} = P(a^2 + c^2)\frac{L}{6} + \left(\frac{R}{2}\right)(4\pi - I), \tag{35}$$

where $P = \frac{3}{4\pi}\frac{(3K^0 + \mu^0)}{(3K^0 + 4\mu^0)}$; $R = \frac{3}{4\pi}\frac{\mu^0}{(3K^0 + 4\mu^0)}$; $L = \frac{(4\pi - 3I)}{(a^2 - c^2)}$.

For $c < a$
$$I = \frac{2\pi a^2 c}{(a^2 - c^2)^{3/2}}\left[\cos^{-1}(c/a) - (c/a^2)\sqrt{(a^2 - c^2)}\right]. \tag{36}$$

For $c > a$
$$I = \frac{2\pi a^2 c}{(c^2 - a^2)^{3/2}}\left[-\cosh^{-1}(c/a) + (c/a^2)\sqrt{(c^2 - a^2)}\right]. \tag{37}$$

Appendix II

Eq. (25) gives

$$v_1\left(2\overline{Q}_{44}\right)_1 + v_2\left(2\overline{Q}_{44}\right)_2 = 1 \tag{38}$$

and
$$v_1\left(\overline{Q_{11} + 2Q_{12}}\right)_1 + v_2\left(\overline{Q_{11} + 2Q_{12}}\right)_2 = 1. \tag{39}$$

Similarly eq. (23) gives

$$v_1(\mu_1 - \mu^*)\left(2\overline{Q}_{44}\right)_1 + v_2(\mu_2 - \mu^*)\left(2\overline{Q}_{44}\right)_2 = 0 \tag{40}$$

and
$$v_1(K_1 - K^*)\left(\overline{Q_{11} + 2Q_{12}}\right)_1 + v_2(K_2 - K^*)\left(\overline{Q_{11} + 2Q_{12}}\right)_2 = 0. \tag{41}$$

From eqs. (38) and (41), eq. (26) follows. Similarly eqs. (38) and (39) gives eq. (27).

Explicit expressions for $\left(\overline{Q_{11} + 2Q_{12}}\right)_1$ and $\left(2\overline{Q}_{44}\right)_1$ are given by (for spheroidal grains)

$$\left(\overline{Q_{11} + 2Q_{12}}\right)_1 = \frac{1}{3}\frac{3 + s_1U}{1 + 3s_3 + s_1\left(\frac{1}{3}U + s_2V\right)} \tag{42}$$

and
$$\left(2\overline{Q}_{44}\right)_1 = \frac{1}{5}\left[\frac{2}{1 + s_1(A_{11} - A_{12})} + \frac{3K_1 + 4\mu^*}{\lambda(3K^* + 4\mu^*)} + \frac{1}{\frac{1}{2} + s_1A_{44}}\right], \tag{43}$$

where
$$U = A_{11} + A_{12} - 2A_{13} - 2A_{31} + 2A_{33},$$

$$V = A_{33}(A_{11} + A_{12}) - 2A_{13}A_{31},$$

$$s_1 = \frac{\mu_1 - \mu^*}{\mu^*}, \quad s_2 = \frac{K_1 - K^*}{K^*}, \quad s_3 = \frac{K_1 - K^*}{3K^* + 4\mu^*},$$

and
$$\lambda = \frac{3K_1 + 4\mu^*}{3K^* + 4\mu^*} + s_1\left(\frac{1}{3}U + s_2V\right).$$

Expressions for $\left(\overline{Q_{11}+2Q_{12}}\right)_1$ and $\left(2\overline{Q}_{44}\right)_1$ are immensely simplified for sphere, needle and disc.

For sphere :

$$a = c, \quad I = \frac{4\pi}{3}, \quad L = \frac{4\pi}{5a^2}.$$

$$A_{11} = A_{33} = 4\pi\left(\frac{Q}{5} + \frac{R}{3}\right),$$

$$A_{12} = A_{13} = A_{31} = \frac{4\pi}{3}\left(\frac{Q}{5} - R\right),$$

$$A_{11} + 2A_{12} = \frac{3K^*}{3K^* + 4\mu^*},$$

$$A_{11} - A_{12} = \frac{6}{5}\frac{K^* + 2\mu^*}{3K^* + 4\mu^*},$$

$$A_{44} = A_{55} = A_{66} = \frac{3}{5}\frac{K^* + 2\mu^*}{3K^* + 4\mu^*},$$

$$U = \frac{18}{5}\frac{K^* + 2\mu^*}{3K^* + 4\mu^*},$$

$$V = \frac{18}{5}\frac{K^*(K^* + 2\mu^*)}{(3K^* + 4\mu^*)^2},$$

$$\lambda = \frac{3K_1 + 4\mu^*}{3K^* + 4\mu^*}\left[1 + \frac{6(\mu_1 - \mu^*)(K^* + 2\mu^*)}{5\mu^*(3K^* + 4\mu^*)}\right].$$

Thus
$$\left(\overline{Q_{11}+2Q_{12}}\right)_1 = \frac{3K^* + 4\mu^*}{3K_1 + 4\mu^*} \tag{44}$$

and
$$\left(2\overline{Q}_{44}\right)_1 = \frac{5\mu^*(3K^* + 4\mu^*)}{5\mu^*(3K^* + 4\mu^*) + 6(\mu_1 - \mu^*)(K^* + 2\mu^*)}. \tag{45}$$

For needle :

$$a = 0, \quad I = 2\pi, \quad L = \frac{2\pi}{c^2}$$

$$A_{33} = 0\,;\; A_{31} = 0.$$

Hence
$$V = 0$$

$$A_{11} = \frac{3}{4}\frac{3K^* + 3\mu^*}{3K^* + 4\mu^*}\,; \qquad A_{12} = \frac{1}{4}\frac{3K^* - 5\mu^*}{3K^* + 4\mu^*}\,;$$

$$A_{13} = \frac{1}{2}\frac{3K^* - 2\mu^*}{3K^* + 4\mu^*}\,; \qquad A_{11} - A_{12} = \frac{3K^* + 7\mu^*}{2(3K^* + 4\mu^*)}\,;$$

$$A_{44} = \frac{1}{4}\,;$$

$$\lambda = \mu_1 + 3(K_1 + \mu^*)\,;$$

$$U = \frac{3\mu^*}{3K^* + 4\mu^*}\,;$$

Therefore, $$\left(\overline{Q_{11}+2Q_{12}}\right)_1 = \frac{\mu_1 + 3(K^* + \mu^*)}{\mu_1 + 3(K_1 + \mu^*)} \tag{46}$$

and $$\left(2\overline{Q}_{44}\right)_1 = \frac{1}{5}\left[\frac{2}{1+s_1 s_4} + \frac{3K_1 + 4\mu^*}{\mu_1 + 3(K_1 + \mu^*)} + \frac{1}{\frac{1}{2}+\frac{s_1}{4}}\right], \tag{47}$$

where $$s_4 = \frac{3K^* + 7\mu^*}{2(3K^* + 4\mu^*)}.$$

For disc :

$$c = 0, \quad l = 0, \quad L = \frac{4\pi}{a^2}$$

$$A_{11} = 0; \quad A_{33} = 4\pi\left(\frac{Q}{3} + R\right);$$

$$A_{12} = 0; \quad A_{13} = 0;$$

$$A_{31} = 4\pi\left(\frac{Q}{3} - R\right); \quad A_{44} = 2\pi\left(\frac{Q}{3} + R\right) = \frac{1}{2};$$

$$U = 2(A_{33} - A_{31}) = \frac{12\mu^*}{3K^* + 4\mu^*};$$

$$V = 0;$$

$$\lambda = \frac{3K_1 + 4\mu_1}{3K^* + 4\mu^*}.$$

Then $$\left(\overline{Q_{11}+2Q_{12}}\right)_1 = \frac{3K^* + 4\mu_1}{3K_1 + 4\mu_1}, \tag{48}$$

$$\left(2\overline{Q}_{44}\right)_1 = \frac{1}{5}\left[\frac{2(\mu_1 + \mu^*)}{\mu_1} + \frac{3K_1 + 4\mu^*}{3K_1 + 4\mu_1}\right]. \tag{49}$$